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Three-body-correlation effect on the ground-state properties of ^{3}He and ^{3}He↑

C. Lhuillier
Laboratoire de Spectroscopie Hertzienne de l'Ecole Normale Supérieure,
24 rue Lhomond, F75231 Paris Cedex 05, France

D. Levesque
*Laboratoire de Physique Théorique et Hautes Energies,**
Université de Paris-Sud, 91405 Orsay Cedex, France

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We report some Monte Carlo computations of the ground-state properties of ^{3}He and ^{3}He↑, using a variational wave function including explicit three-body correlations. This inclusion results in a slight improvement of the computed properties: internal energy, equilibrium density, and compressibility. The comparison with experiments on ^{3}He remains still largely unsatisfactory.

I. INTRODUCTION

Until very recently the most accurate variational description of liquid ^{3}He was that obtained with a wave function of the Jastrow-Slater (JS) type

$$\Psi_{\rm JS}=\left(\prod_{i<j}\chi_{ij}\right)\Phi \ , \tag{1}$$

where χ_{ij} is a two-body function which essentially prevents the overlap of the cores of the neighboring particles, while Φ is the solution of the free-fermion problem, that is, a Slater determinant of plane waves with wave vectors filling the Fermi sea, for each "species" of ^{3}He with up and down spins. Some of the failures of the Jastrow wave functions are quite well known: in this model the equilibrium density is 20% too low (13×10^{-3} Å^{-3} as compared with the experimental value of 16×10^{-3} Å^{-3}), the compressibility too high (the results of Ref. 1 lead to a compressibility under zero pressure approximately 15% too high), and the fluid so described lacks local order. Moreover this modeled fluid is a spontaneous ferromagnet; that is, ^{3}He↑ (spin-polarized ^{3}He) appears to be more stable than ordinary ^{3}He in the whole domain of existence of the liquid. As discussed in Refs. 1 and 2 this "spontaneous ferromagnetism" could be a mere consequence of the failure of Eq. (1) to give a correct localization of the nearest neighbors. So it was interesting to try to improve on Eq. (1) to have a better structural description of liquid ^{3}He. The study of the local order exhibited by the helium liquids is a great help in achieving this task. The x-ray spectrum of helium[3,4] strongly suggests that the *local* order of liquid helium is best described by a configuration of the T_d^2 space group (i.e., a face-centered-cubic lattice in which half of the atoms are removed; see Fig. 1). Such a local arrangement is in very good agreement with the position of the first peak of the radial distribution function (six nearest neighbors at a distance of 3.16 Å in ^{3}He). This hypothesis elaborated by Keesom and Taconis[3] has been thoroughly studied by London.[5] The good agreement between that very simple model of a quasisolid and the experimental results in the whole domain of existence of the liquid is impressive and is a strong argument in favor of a statistical preference for such a local arrangement. If we analyze such an arrangement with the help of the simplest structure, we see (Fig. 1) that it is built up from regular tetrahedrons, that is fundamentally from equilateral configurations.

The need for stabilization of equilateral configurations has been recognized for a long time. The first attempt has been made for ^{4}He by de Michelis and

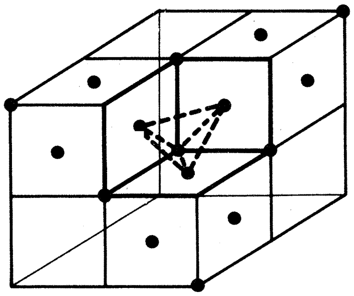

FIG. 1. T_d^2 lattice of Keesom and Taconis.

Reatto[6] who tried to improve on the usual Jastrow factor

$$\chi(r)=\exp[-u(r)]$$

with

$$u(r)=\frac{1}{2}\left(\frac{b}{r}\right)^m \tag{2}$$

by modifying the two-body function $u(r)$ to incorporate in it some medium-range attractive effect and a repulsive pseudopotential in order to simulate three-body effects. This attempt led to an improvement on the two-body correlation factor, but the improvement on the binding energy was not really significant; this could be attributed to the low efficiency of a two-body correlation to stabilize equilateral configurations.

Again in the case of ^{4}He, Woo and Coldwell[7] introduced a repulsive three-body correlation. The calculation was made in two dimensions, the improvement was small but significant.

More recently, Schmidt and Pandharipande[8] (SP) made a calculation for ^{3}He with a new wave function based on the Feynman-Cohen correlation operator. This calculation leads to a large improvement in the binding energy at equilibrium density but the compressibility of the fluid so described was twice as large as it should be. Moreover these calculations were done in the framework of Fermi-hypernetted-chain (FHNC) techniques which do not ensure the upper-bound property of the variational energy.

Using a somewhat simpler wave function, including only the three-body correlations used by SP and no momentum-dependent correlations, we have now performed a Monte Carlo computation for both the unpolarized and totally polarized phase of liquid ^{3}He. In Sec. II we describe the relevant features of this new wave function and we briefly discuss the choice of the interatomic potential. The internal energy and the two-body correlation functions have been computed through an exact Monte Carlo algorithm first proposed by Ceperley *et al.*[9] The specific details of the present computation are given in the Appendix. In Sec. III we give the results of these computations and compare both with experiment and with results obtained in a pure Jastrow model.

II. VARIATIONAL MODEL OF LIQUID HELIUM

A. New wave function

The wave function we used is of the following form:

$$\Psi=\Psi_{JS}\prod_{i<j<k}\zeta(i,j,k)\ , \tag{3}$$

where Ψ_{JS} is given by Eq. (1). The three-body correlation is of the following form:

$$\prod_{i<j<k}\zeta(i,j,k)=\exp\left(\sum_{i<j<k}\ \sum_{\text{cycl}}\xi(r_{ij})\xi(r_{ik})\vec{r}_{ij}\cdot\vec{r}_{ik}\right)\ ; \tag{4}$$

where $\sum_{\text{cycl}}$ means sum upon the cyclic permutation of the three indices i,j,k, with

$$\xi(r)=\beta\frac{1}{2}m\frac{b^m}{r^{m+2}}\exp\left[-\left(\frac{1}{r}\right)^m\right]\ ; \tag{5}$$

b and m are the variational parameters of the two-body correlation given by Eq. (2) (here m is given the value 5) and $\vec{r}_{ij}=\vec{r}_i-\vec{r}_j$. Unless otherwise specified, all the distances are in unit σ, the scaling length of the Lennard-Jones potential.

This new wave function differs from the Jastrow-Slater wave function by a medium-range three-body correlation. Its main advantage is that it favors equilateral configuration at a distance of the order of σ (the scaling range of the potential). Moreover, taking an equilateral configuration the three-body factor will strongly resist a deformation of type *a* (Fig. 2) (the correlation becomes repulsive for $\theta>\theta_0$, $\theta_0\sim 90°$). Unfortunately it will not strongly prevent a deformation of type *b* (Fig. 2), which is probably a very serious drawback of the angular form given by Eq. (4). The radial part of this correlation [Eq. (5)] has been roughly modeled on that of SP. For computational convenience we chose an exponential form in Eq. (4) instead of the linear form of SP. So the form of ξ has to be somewhat different from that of SP in order to eliminate some undesirable configurations of type *b* (Fig. 2) where the weight of the attractive three-body correlation could partially cancel the repulsive two-body correlation. The only variational parameter is the weight β of the three-body correlation.

B. Interatomic potential

The features of the wave function being fixed, a new choice must be made: that of the potential. The

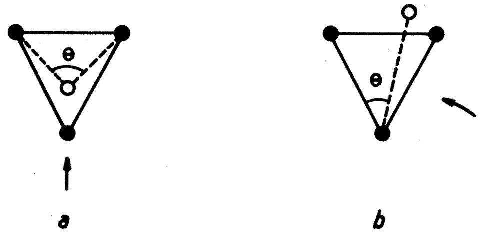

FIG. 2. Some typical deformations of the triangular configurations.

TABLE I. Numerical results for the energy of the unpolarized phase. ρ is the density (number of particles by σ^3), β the weight of the three-body correlation, b is the parameter of the two-body function, E_k is the kinetic energy of the particles, $E_p(1)$ is the potential energy computed with the Lennard-Jones potential, $E_p(2)$ is the potential energy computed with the HFDHE2 potential of Aziz *et al.* (Ref. 11), E_{tot} is the total internal energy by particles (all energies in K), n is the number of configurations sampled in the MC run ($\times 10^4$). For all these results the computation was made on a sample of 54 particles with periodic boundary conditions, and the error estimate is of the order of 0.15 K. $[E_{\text{tot}} = E_k + E_p(1)]$.

ρ	β	b	E_k	$E_p(1)$	$E_p(2)$	E_{tot}	n
0.414	0.25	1.145	22.91	−21.06	· · ·	1.85	43.2
· · ·	0.25	1.145	23.00	−20.96	−20.18	2.04	43.2
· · ·	0.35	1.145	22.65	−20.63	· · ·	2.02	43.2
0.3648	0.25	1.12	17.97	−17.65	−17.02	0.32	43.2
0.3283	0.25	1.12	15.56	−16.00	−15.67	−0.44	43.2
0.30	0.25	1.12	13.52	−14.70	−14.46	−1.18	43.2
0.2743	0.25	1.12	11.93	−13.40	· · ·	−1.47	30.0
· · ·	0.25	1.12	12.01	−13.32	−13.29	−1.31	43.2
· · ·	0.35	1.145	12.67	−13.91	−14.13	−1.24	43.2
· · ·	0.35	1.17	13.42	−14.30	−14.71	−0.88	43.2
0.2554	0.25	1.12	10.84	−12.36	−12.38	−1.52	43.2
0.236	0.25	1.12	9.98	−11.23	−11.26	−1.25	43.2
· · ·	0.25	1.10	9.45	−10.83	−10.76	−1.38	43.2

interaction between the ^{3}He atoms is usually described by the Lennard-Jones potential

$$V(r) = 4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right],$$

with $\epsilon = 10.22$ K and $\sigma = 2.556$ Å. This potential is known to slightly underbind the liquid ^{4}He at the lower densities.[10] To have an indication of the sensitivity of the results to the potential, we have tried another realistic potential which has been recently proposed by Aziz *et al.*[11] This new potential (HFDHE2) presently gives the best account of the gaseous properties of helium over a very large

TABLE II. Results for the polarized phase. Same notation as in Table I. (The size of the sample is now of 57 particles.)

ρ	β	b	E_k	$E_p(1)$	$E_p(2)$	E_{tot}	n
0.414	0.25	1.13	22.60	−21.54	· · ·	1.16	45.6
· · ·	0.35	1.13	22.36	−21.26	−20.79	1.10	45.6
· · ·	0.45	1.13	22.88	−20.74	· · ·	2.14	45.6
0.3648	0.25	1.13	19.23	−19.27	· · ·	−0.04	68.4
· · ·	0.25	1.12	18.96	−18.96	−18.69	0.00	45.6
· · ·	0.25	1.10	17.85	−17.91	· · ·	−0.06	45.6
· · ·	0.35	1.13	19.21	−19.33	· · ·	−0.12	45.6
0.3283	0.25	1.13	16.42	−17.18	−17.32	−0.76	36.7
· · ·	0.25	1.12	15.85	−17.03	· · ·	−1.18	45.6
0.30	0.25	1.12	13.98	−15.48	−15.68	−1.50	45.6
0.2743	0.25	1.12	12.43	−13.99	· · ·	−1.56	45.6
· · ·	0.25	1.10	12.00	−13.55	−13.66	−1.55	45.6
0.2554	0.25	1.12	11.37	−12.83	−13.12	−1.46	45.6
· · ·	0.25	1.10	11.12	−12.44	· · ·	−1.32	45.6
0.236	0.25	1.10	9.94	−11.42	−11.61	−1.48	45.6
0.2189	0.25	1.10	9.08	−10.36	−10.57	−1.28	45.6

domain of temperature and energy. It differs from the Lennard-Jones potential by the size of the repulsive core, which is a bit larger, and of the well depth (10.8 vs 10.22 K).

III. RESULTS

The expectation values of the Hamiltonian and two-body correlation function have been computed through an exact Monte Carlo algorithm. The method of computation has been thoroughly described in Refs. 1 and 9; the relevant formulas for this specific problem are given in the Appendix. The computations have been made on samples of 54–57 particles. The number of generated configurations is of the order of 450×10^3 for each run, which ensures a statistical uncertainty of the order of 0.15 K.

The results are reported in Tables I and II, and Figs. 3–6. We will begin the discussion by the analysis of the two-body correlation function which sheds light on the other results.

A. Two-body correlation function $g(r)$

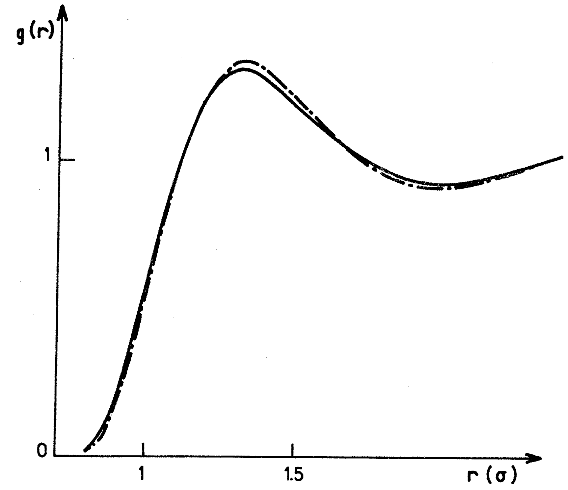

FIG. 4. Two-body correlation functions near the solidification line ($\rho=0.414\sigma^{-3}$) are shown on the figure: —— solid line, $g(r)$ for ordinary ^{3}He; —·— dotted-dashed line, $g(r)$ for ^{3}He↑.

The introduction of the three-body correlation improves only slightly the two-body correlation function (Figs. 3 and 4). At the equilibrium density of real ^{3}He ($\rho=0.274\sigma^{-3}$), the improvement in local structure is slight for the unpolarized phase and insignificant for the polarized phase. This different behavior can easily be understood by the fact that the statistical correlations induced by the Slater determinants are more dominant in the polarized case than in the unpolarized one. For the unpolarized phase the new wave function does not yet give enough local order to match the experimental results.[4]

B. Internal energy

The introduction of the three-body correlation improves the properties of the unpolarized phase significantly, essentially at high density (Figs. 5 and 6). The variation of the internal energy with the

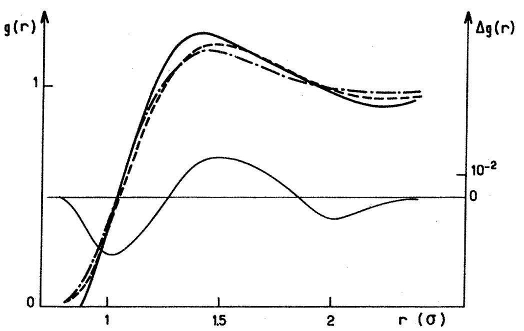

FIG. 3. Two-body correlation functions at equilibrium density of real ^{3}He ($\rho=0.274\sigma^{-3}$) are shown in the figure: —— solid line, the experimental results of Achter and Meyer (Ref. 4); —·— dotted-dashed line, the computational results for ordinary ^{3}He described by Eq. (3); – – – dashed line, the results for ^{3}He↑ given by Eq. (3). On the same figure (scale on the right) we report the improvement in $g(r)$ induced by the use of explicit three-body correlations for ordinary ^{3}He. $\Delta g(r)=g(\mathrm{Eq.}(3))-g(\mathrm{Eq.}(1))$.

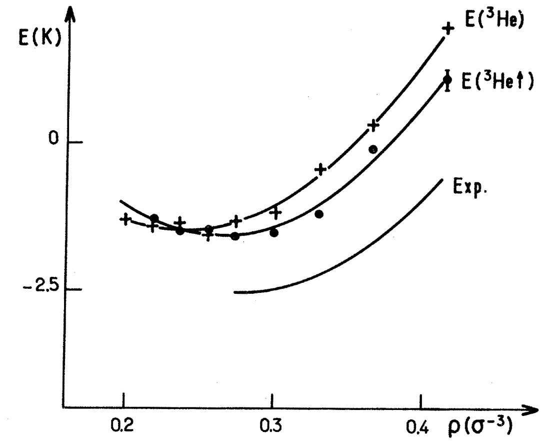

FIG. 5. Internal energy of ^{3}He and ^{3}He↑ computed with the help of Eq. (3). The lower solid line represents the experimental results for ordinary ^{3}He [extrapolated from the results of Boghosian *et al.* (Ref. 13)]. The typical statistical uncertainty upon a MC run is shown on the right.

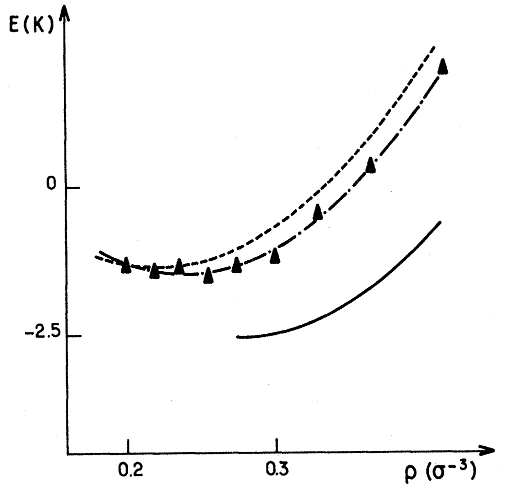

FIG. 6. Internal energy of ordinary ^{3}He. The dotted curve represents the results obtained from the pure Jastrow-Slater wave function (Ref. 1); the dotted-dashed curve, the results obtained with the inclusion of three-body correlations (this work); the solid line, the experimental results.

density can be described by a polynomial expansion

$$E=E_0+A\left(\frac{\rho-\rho_0}{\rho_0}\right)^2+B\left(\frac{\rho-\rho_0}{\rho_0}\right)^3+\cdots .$$

We fitted our results with the first two terms (the variance of the results being not significantly lowered by the introduction of the cubic term). The results for the wave function Eq. (3) lead to the following estimates for the unpolarized phase (in parentheses are given the results for a pure Jastrow function from Ref. 1):

	Pure Jastrow
$E_0=-1.46\pm0.04$ K	$(E_0=-1.35\pm0.05$ K),
$\rho_0=0.236\pm0.006\sigma^{-3}$	$(\rho_0=0.22\pm0.007\sigma^{-3}$),
$A=5.8\pm0.7$ K	$(A=5.1\pm0.7$ K).

The improvement in equilibrium density is small but significant and the improvement in compressibility is noticeable.

The results for the polarized phase (^{3}He↑) are the following:

	Pure Jastrow
$E_0=-1.56\pm0.06$ K	$(E_0=-1.55\pm0.03$ K),
$\rho_0=0.265\pm0.006\sigma^{-3}$	$(\rho_0=0.259\pm0.003\sigma^{-3}$),
$A=8.7\pm1.2$ K	$(A=7.6\pm0.6$ K).

The improvement on the pure Jastrow wave function appears to be quite insignificant in the polarized case. We have already underlined this phenomenon in the case of the two-body correlation function and explained it by the weight of the statistical correlations. This argument can even be stated in a more general way. The symmetry requirement is much more stringent for a totally polarized fluid than for an unpolarized one, which could explain why the polarized wave function is much better than the unpolarized one. In passing we note that the numerical results support the idea that ^{3}He↑ is less compressible than ordinary ^{3}He. This feature seems to us a reasonable prediction, the reinforcement of the Pauli principle in ^{3}He↑ should give rise to a stronger localization of the particles and reinforce the quasisolid behavior of ^{3}He↑.

All the numerical results given above are relative to the Lennard-Jones potential. In Tables I and II we have quoted some results relative to the HFDHE2 potential.[11] These results are partial ones: in particular the parameter *b* of the two-body correlation would need to be adjusted for this new potential. This adjustment has been made at the equilibrium density of liquid ^{3}He ($\rho=0.274\sigma^{-3}$). The results obtained are a bit lower than those obtained with Lennard-Jones potential, but the lowering is really very small and does not notably modify the picture: in particular this modification of the potential is not sufficient to solve the paradox of the "ferromagnetism" of our model fluid: a paradox which probably lies more in the inadequacy of the variational wave function than in a failure of the interatomic potential.

In conclusion, the introduction of three-body correlations of the kind described above does not lead to large improvements of the structural properties of liquid ^{3}He. The equilibrium density and compressibility of the fluid are slightly improved with regard to the pure Jastrow model and the artificial ferromagnetism of the fluid is somewhat diminished. It is possible that a greater flexibility in the form of the radial part could lead to some lowering of the variational energy. Nevertheless a preliminary investigation led us to think that such an improvement could not qualitatively modify the two main drawbacks of the picture: the discrepancy of about 1 K in the internal energy at equilibrium and the paradox of the ferromagnetism of the ground state. The comparison with the results obtained[12] in the same approach for ^{4}He puts the emphasis on the fact that some essential physical feature of ^{3}He must be lacking in the wave function given by Eq. (3).

APPENDIX

In this appendix we do not repeat the contribution to the kinetic energy of the Slater determinants. The reader is referred to Refs. 9 or 1. We just emphasize the problem of computation associated with the three-body factor. First for computational convenience the symmetric part of the wave function:

$$\Psi_S = \left(\prod_{i<j} \chi_{ij}\right)\left(\prod_{i<j<k} \zeta(i,j,k)\right) \tag{A1}$$

could be written in the form

$$\Psi_S = \exp\left(-\tfrac{1}{4} \sum_{i \neq j} (u_{ij} + 2v_{ij}) + \tfrac{1}{2} \sum_{i} \vec{F}_i^{\,2}\right) \tag{A2}$$

with

$$u_{ij} = \left(\frac{b}{r_{ij}}\right)^m , \quad v_{ij} = \xi_{ij}^2 r_{ij}^2 , \quad \vec{F}_i = \sum_{j \neq i} \xi_{ij} \vec{r}_{ij} .$$

We choose to use the following form of the kinetic energy (which is readily obtained from the more classical form by use of the Green's theorem)

$$T = \frac{h^2}{2m_0} \sum_m \langle \vec{\mathfrak{F}}_m^{\,2} \rangle \; ;$$

m_0 is the mass of ^{3}He atom and

$$\vec{\mathfrak{F}}_m = \frac{\vec{\nabla}_m \Psi}{\Psi} .$$

The contribution of the Slater determinants $\vec{\mathfrak{F}}_m(\mathrm{det})$ is the same as in the previous computations (see Ref. 1). The new term can be written

$$\begin{aligned} \vec{\mathfrak{F}}_m - \vec{\mathfrak{F}}_m(\mathrm{det}) = \tfrac{1}{2} \sum_{j \neq m} (u' + 2v') \frac{\vec{r}_{mj}}{r_{mj}} \\ + \sum_k (\vec{F}_m - \vec{F}_k) \\ \times \left(\xi_{mk} + \xi'_{mk} \cdot \frac{\vec{r}_{mk} \vec{r}_{mk}}{r_{mk}}\right) \end{aligned} \tag{A3}$$

(where the prime indicates the derivative of the function with respect to r, the interparticle distance).

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[*] Laboratoire associe au Centre National de la Recherche Scientifique.

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